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OPTICS AND SPECTROSCOPY

DESCRIPTION OF EVAPORATION OF A SPHERICAL LIQUID DROP BY A NON-MARKOVIAN RANDOM PROCESS BASED ON INTEGRAL STOCHASTIC EQUATIONS

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Processes of evaporation (condensation) of vapor particles from the surface of a spherical drop and processes of their diffusion into surrounding volume are considered. Special features of evaporation are investigated taking into account vapor particle fluctuations caused by random changes in the temperature, concentration, etc. Statistical characteristics of fluctuations of the corresponding quantities, including the mass flow through the liquid-vapor boundary and concentration on the liquid surface, are presented. The distribution of completely evaporated drop number versus time is presented.

Keywords: aerosol particles, diffusion, non-Markovian processes.

Processes of evaporation (condensation) of vapor particles from (on) the surface of a liquid aerosol drop and diffusion of these particles into the surrounding atmosphere are intensively studied. This is due to widespread distribution of substances in the state of the liquid-drop aerosol and their practical importance. By way of examples, we note that many objects (both of natural and technogenic origin) that can be met in the atmosphere have the above-indicated character [1]; the aerosols are natural catalysts of photochemical and other reactions in the atmosphere and end products of a number of processes. Their study is important for meteorological monitoring (for example, for weather forecasting), ecological investigations (in particular, for the determination of concentration of poisonous substances in the atmosphere), etc. The problem of aerosol formation is urgent because of intensive investigations of plasma chemical processes in a gas discharge [2].

Many biological objects that can be met in nature and in living organisms are liquid-drop aerosols [3]. In some cases, when performing biological and medical studies, the behavior of such aerosols and particles evaporated from their surfaces must be considered.

The diffusion processes are used in chemical kinetics and engineering to regulate the course of chemical reactions and to separate substances [4]. During chemical reactions, one (or several) reacting substances can be in the state of liquid-drop aerosol; in this regard, the processes of particle evaporation from the spherical drop surface must be studied for more accurate control over the reaction course.

We note that the processes of evaporation (condensation) of liquid particles are always accompanied by random fluctuations of temperature at the boundary between the liquid and gaseous phases, concentration of vapor particles at the drop surface, etc. Such fluctuations are disregarded in conventional description of drop evaporation. In this case, the diffusion processes can be studied by the well-developed classical methods using differential operators, and the Markovian evaporation (condensation) and diffusion processes are considered [5].

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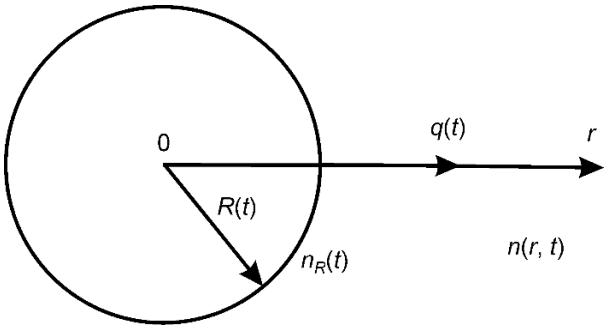

Fig. 1. Vapor diffusion over the spherical particle surface.

However, the Markov theory is only the first approximation. Thus, in [6] it was demonstrated that consideration of entrainment by the Brownian particle of the surrounding particles calls for the application of integral equations and the theory of non-Markovian processes.

In the present work, processes of evaporation of a spherical liquid drop and diffusion of vapor particles are considered with allowance for the above-indicated fluctuations. As demonstrated below, this calls for the application of stochastic integral equations and hence theories of non-Markovian random processes [7].

PROBLEM FORMULATION

Let us consider a fixed spherical liquid drop with initial radius R_0 around which diffusion of vapor particles occurs (Fig. 1). The origin of the spherical system of coordinates is placed in the drop centre. We designate by n_0 the concentration of saturated vapors of the liquid drop and consider that the concentration of vapor at the liquid drop surface is a function of time designated by $n_R(t)$. We assume also that the diffusion coefficient D is constant. The mass of one liquid particle is designated by m. We consider further that the atmosphere surrounding the drop is completely saturated with liquid vapor.

Obviously, with allowance for the symmetry of the examined problem, the vapor particle concentration depends only on the distance to the drop centre r and time t. The diffusion equation in the spherical coordinates in this case assumes the form

$$\frac{\partial n(r,t)}{\partial t} = \frac{D}{r}\frac{\partial^2 (rn(r,t))}{\partial r^2}. \tag{1}$$

As indicated above, the boundary and initial conditions are written with the help of expressions

$$n(r,t)\big|_{r=R} = n_R(t), \tag{2}$$

$$n(r,t)\big|_{t=0} = n_0. \tag{3}$$

We note that the particle radius R in condition (2) is generally time-dependent.

Evaporation and condensation of vapor occur on the boundary of the liquid. It is clear that particle condensation is observed when the concentration of vapor particles at the surface $n_R(t)$ exceeds the concentration of saturated vapor n_0. Otherwise, evaporation of the vapor particles from the drop surface is observed.

Taking the direction along the coordinate r for the positive direction of flow of vapor particles from the surface $q(t)$ and considering that this flow is proportional to the difference between concentrations $n_R(t)$ and n_0, we obtain

$$q(t) = -\alpha\left(n_R(t) - n_0\right) + \xi_q(t)\,. \tag{4}$$

In the last expression, we have considered the above-indicated fluctuations of the vapor particle flow described by the last term $\xi_q(t)$. The characteristic coefficient α in Eq. (4), often called the diffusion accommodation coefficient, depends generally on the temperature, species of the substance etc. and has the dimensionality of velocity.

Obviously, the particle flow $q(t)$ obeys the general formula

$$q(t) = -D\left.\frac{\partial n}{\partial r}\right|_{r=R}. \tag{5}$$

However, it would be more vividly to consider the mass flow $q_m(t)$ equal to the mass of particles evaporated (condensed) from (on) the unit area per unit time rather than the particle flow through the drop surface $q(t)$. It is easy to understand that the corresponding expression for the mass flow $q_m(t)$ is derived by multiplication of Eqs. (4) and (5) into the mass of one particle m. In other words,

$$q_m(t) = -m\alpha\left(n_R(t) - n_0\right) + \xi_{q_m}(t)\,, \tag{6}$$

$$q_m(t) = -mD\left.\frac{\partial n}{\partial r}\right|_{r=R}. \tag{7}$$

In Eq. (6), $\xi_{q_m}(t) = m\xi_q(t)$.

Equating Eqs. (6) and (7), we obtain

$$-mD\left.\frac{\partial n}{\partial r}\right|_{r=R} = -m\alpha\left(n_R(t) - n_0\right) + \xi_{q_m}(t)\,. \tag{8}$$

The intensity of mass flow fluctuations σ and the value of the coefficient α can be estimated using the characteristic problem parameters (the saturated vapor concentration n_0, temperature T, liquid particle mass m, and diffusion coefficient D) from the following expressions:

$$\sigma = Dm^2 n_0^2\,, \tag{9}$$

$$\alpha = \sqrt{\frac{kT}{m}}\,, \tag{10}$$

where k is the Boltzmann constant.

QUASISTATIONARY-CASE

Let us consider first the problem under assumption of constancy of particle radius: $R(t) = R_0$. This approach is physically substantiated for large particle radius and relatively short observation time of diffusion processes during which changes in the drop sizes can be neglected.

Let us solve system of equations (1)–(3). For this purpose, we introduce the auxiliary function $F(r,t)$ specified by the formula

$$n(r,t) = \frac{F(r,t)}{r}. \tag{11}$$

Substitution of the last formula into Eqs. (1) – (3) yields the following system of equations for the function $F(r,t)$:

$$\frac{\partial F(r,t)}{\partial t} = D\frac{\partial^2 F(r,t)}{\partial r^2}, \tag{12}$$

$$F(r,t)\big|_{r=R} = R_0 n_R(t), \tag{13}$$

$$F(r,t)\big|_{t=0} = n_0 r, \quad r > R_0. \tag{14}$$

From Eqs. (12)–(14) it follows that the function $F(r,t)$ corresponds formally to the one-dimensional diffusion equation. Therefore, a solution of the formulated problem has the form [8]

$$F(r,t) = \frac{R_0}{2\sqrt{\pi D}}\int_0^t \frac{r-R_0}{(t-\tau)^{3/2}}\exp\left[-\frac{(r-R_0)^2}{4D(t-\tau)}\right] n_R(\tau)d\tau, \ r > R_0. \tag{15}$$

We note that in Eq. (15) it is considered that dynamic equilibrium between the flow determined by the initial concentration of vapor particles and the flow of evaporating liquid is established almost instantaneously; therefore, the term caused by the presence of the initial concentration has been omitted from Eq. (15).

Let us now calculate the derivative $\left.\frac{\partial F(r,t)}{\partial r}\right|_{r=R}$ with allowance for Eq. (7). We obtain

$$\left.\frac{\partial F(r,t)}{\partial r}\right|_{r=R} = -\frac{R_0}{mD}q_m(t) + n_R(t). \tag{16}$$

The derivative of the last expression with respect to the coordinate *r* is determined from Eq. (15):

$$\frac{\partial F(r,t)}{\partial r} = \frac{R_0}{2\sqrt{\pi D}}\int_0^t \left[\frac{1}{(t-\tau)^{3/2}} - \frac{2(r-R_0)^2}{4D(t-\tau)^{5/2}}\right]\exp\left[-\frac{(r-R_0)^2}{4D(t-\tau)}\right] n_R(\tau)d\tau. \tag{17}$$

Calculations of integral (17) by parts yields

$$\frac{\partial F(r,t)}{\partial r} = -\frac{R_0}{\sqrt{\pi D}}\int_0^t \frac{1}{\sqrt{t-\tau}}\exp\left[-\frac{(r-R_0)^2}{4D(t-\tau)}\right]\frac{dn_R(\tau)}{d\tau}d\tau. \tag{18}$$

Comparing Eqs. (16) and (17), we obtain expression for the mass flow $q_m(t)$:

$$q_m(t) = \frac{mD}{R_0}\int_0^t \left(\frac{R_0}{\sqrt{\pi D}}\frac{1}{\sqrt{t-\tau}} + 1\right)\frac{dn_R(\tau)}{d\tau}d\tau. \tag{19}$$

Let us introduce the substitutions

$$\delta n(t) = n_R(t) - n_0, \tag{20}$$

$$Z(t) = \frac{d\delta n(t)}{dt}. \tag{21}$$

With allowance for Eqs. (20) and (21), from Eqs. (6) and (19) we finely obtain

$$m\int_0^t \left(\sqrt{\frac{D}{\pi}} \frac{1}{\sqrt{t-\tau}} + \alpha + \frac{D}{R_0} \right) Z(\tau) d\tau = \xi_{q_m}(t). \tag{22}$$

Thus, the random process $Z(t)$ is described by the first-order linear integral Volterra operator whose kernel is the sum of the Abel-type term, a constant, and a quantity depending on the spherical particle radius. The description of the random process $Z(t)$ by integral operator (22) demonstrates the non-Markovian character of the process [7] and hence the application of the theory of non-Markovian processes is required to study this process. We also indicate that the quantity $\delta n(t)$ is generally a non-Markovian random process, because it is represented by the integral over time of the non-Markovian process $Z(t)$.

We note that formula (22) allows the processes of vapor diffusion into the semi-infinite space above the flat liquid surface to be investigated. Indeed, in this case we formally set $R_0 = \infty$. Then from Eq. (22) we obtain the expression

$$m\int_0^t \left(\sqrt{\frac{D}{\pi}} \frac{1}{\sqrt{t-\tau}} + \alpha \right) Z(\tau) d\tau = \xi_{q_m}(t). \tag{23}$$

Let us take the Laplace transform of Eq. (22). We obtain

$$\left(\sqrt{\frac{D}{p}} + \frac{\alpha}{p} + \frac{D}{R_0 p} \right) m\hat{Z}(p) = \hat{\xi}_{q_m}(p), \tag{24}$$

where p is a parameter.

From the last formula we obtain

$$\hat{Z}(p) = \frac{p}{m\left(\sqrt{Dp} + \beta\right)} \hat{\xi}_{q_m}(p), \tag{25}$$

where we have used the substitution

$$\beta = \alpha + \frac{D}{R_0}. \tag{26}$$

Taking advantage of definition (21), we obtain

$$\delta\hat{n}(p) = \frac{1}{m\left(\sqrt{Dp} + \beta\right)} \hat{\xi}_{q_m}(p). \tag{27}$$

Since the integral on the left side of Eq. (22) is a convolution, we seek a solution for $\delta n(t)$ also in the form of the first-order integral Volterra operator having the form of convolution:

$$\delta n(t) = \int_0^t K(t-\tau)\xi_{q_m}(\tau)d\tau\,. \tag{28}$$

Here $K(t-\tau)$ is the thought-after function.

Because the mass flow $q_m(t)$ and $\delta n(t)$ are related by Eq. (6), from Eq. (28) we obtain

$$q_m(t) = \int_0^t Y(t-\tau)\xi_{q_m}(\tau)d\tau\,, \tag{29}$$

where

$$Y(t-\tau) = \delta(t-\tau) - \alpha m K(t-\tau)\,. \tag{30}$$

Taking the Laplace transform of Eq. (28) and equating the expression obtained $\delta\hat{n}(p)$ to Eq. (27), we obtain for the kernel of integral (28) $\hat{K}(p)$:

$$\hat{K}(p) = \left(m\left(\sqrt{Dp} + \beta\right)\right)^{-1}. \tag{31}$$

Taking now the inverse Laplace transform of Eq. (31) (see p. 210 of [9]), we obtain

$$K(t-\tau) = \frac{1}{m\sqrt{D}}\left\{\frac{1}{\sqrt{\pi(t-\tau)}} - \frac{\beta}{\sqrt{D}}\exp\left[\frac{\beta^2(t-\tau)}{D}\right]\operatorname{erfc}\left[\frac{\beta\sqrt{t-\tau}}{\sqrt{D}}\right]\right\}, \tag{32}$$

where $\operatorname{erfc}(x)$ is the complement of the error function [10]:

$$\operatorname{erfc}(x) = \frac{2}{\sqrt{\pi}}\int_x^\infty e^{-s^2}ds\,. \tag{33}$$

By virtue of the fact that with increasing *x*, the function $\operatorname{erfc}(x)$ decreases faster than $\exp(x^2)$, the kernel $K(t-\tau)$ vanishes when $(t-\tau)\to\infty$:

$$K(t-\tau)\big|_{(t-\tau)\to\infty} = 0\,. \tag{34}$$

The plot of the function $K(t-\tau)$ is schematically shown in Fig. 2. In this case, the particle mass *m*, diffusion coefficient *D*, parameter α, and spherical drop radius R_0 were set equal to unity. This is due to the fact that the fast increase of the exponent in Eq. (28) and simultaneous very fast decrease of the function $\operatorname{erfc}(x)$ have complicated calculations with the use of actual parameters. However, it is obvious that for such parameters, the behavior of the function $K(t-\tau)$ will be similar.

The dependence obtained can be approximated with good accuracy by a power-law function. For $(t-\tau) > 0.05$, the function $K(t-\tau)$ is inversely proportional to the difference ($t-\tau$):

$$K(t-\tau) = \frac{0.06}{t-\tau}\,. \tag{35}$$

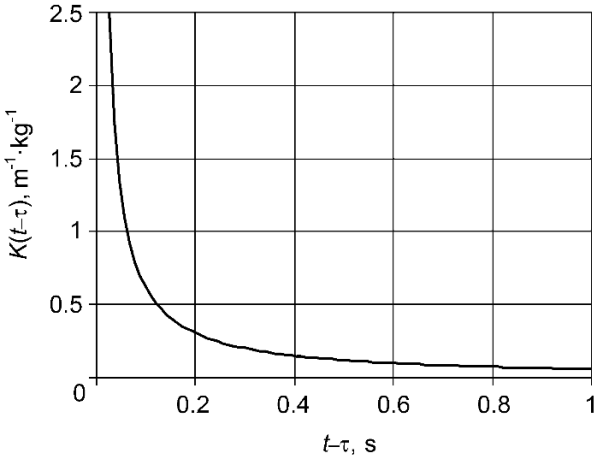

Fig. 2. Plot of the function $K(t-\tau)$ determined by Eq. (32).

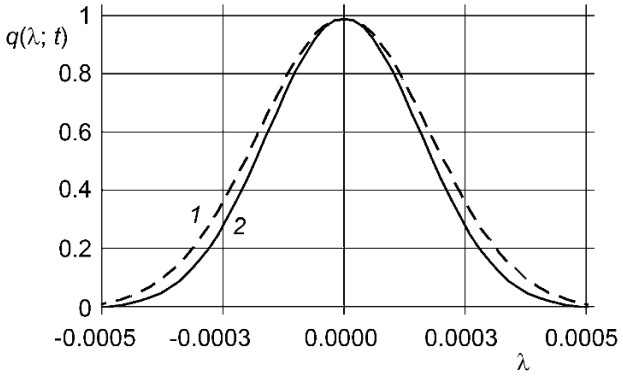

Fig. 3. Plot of the function $g_1(\lambda;t)$ for $t = 5\cdot10^{-6}$ (curve *1*) and 1 s (curve *2*).

Taking advantage of the method described in [11], for the one-dimensional function $g_1(\lambda;t)$ and L-dimensional characteristic functions $g_L(\lambda_1,...,\lambda_L;t_1,...,t_L)$ of the random process $\delta n(t)$, we obtained the following expressions:

$$g_1(\lambda;t) = \exp\left[-\frac{1}{2}\sigma\lambda^2\int_0^t K^2(t-\tau)d\tau\right], \tag{36}$$

$$g_L(\lambda_1,...,\lambda_L;t_1,...,t_L)$$
$$= \exp\left[-\frac{1}{2}\sigma\left(\sum_{k=1}^{L}\lambda_k^2\int_0^{t_k}K^2(t_k-\tau)d\tau + 2\sum_{\substack{k,l=1,\\ k<l}}^{L}\lambda_k\lambda_l\int_0^{t_k}K(t_k-\tau)K(t_l-\tau)d\tau\right)\right]. \tag{37}$$

The plot of the function $g_1(\lambda;t)$ versus t is shown schematically in Fig. 3. From the figure it can be seen that the one-dimensional characteristic function $g_1(\lambda;t)$ is described by the Gaussian curve whose width decreases with increasing time t.

The calculated characteristic functions allow any arbitrary statistical characteristics of the random process $\delta n(t)$ to be calculated. In particular, for its mathematical expectation $\langle \delta n(t) \rangle$ and second-order moment $\langle \delta n(t_1)\delta n(t_2) \rangle$, we have the following expressions:

$$\langle \delta n(t) \rangle = \left. \frac{\partial g_1(\lambda; t)}{i\partial\lambda} \right|_{\lambda=0} = 0, \tag{38}$$

$$\langle \delta n(t_1)\delta n(t_2) \rangle = \left. \frac{\partial^2 g_2(\lambda_1, \lambda_2; t_1, t_2)}{i\partial\lambda_1 \partial\lambda_2} \right|_{\substack{\lambda_1=0, \\ \lambda_2=0}} = \sigma \int_0^{t_1} K(t_1 - \tau) K(t_2 - \tau) d\tau. \tag{39}$$

In Eq. (39), we have considered that $t_1 \le t_2$.

The last formula allows the variance of the process $\delta n(t)$ designated by $\Delta(t)$ to be calculated. In this case, $t = t_1 = t_2$; therefore,

$$\Delta(t) = \langle \delta n^2(t) \rangle = \sigma \int_0^t K^2(t - \tau) d\tau. \tag{40}$$

The statistical characteristics of fluctuations of the particle mass flow $q_m(t)$ are set obviously by formulas similar to Eqs. (36)–(40) in which the kernel $K(t - \tau)$ is replaced by the kernel $Y(t - \tau)$.

We note that integration in the above integrals is performed in the limits from zero to $t - \delta t$, where δt is a small positive parameter equal by the order of magnitude to the free run time of the vapor particles. Introduction of this parameter is physically justified by the fact that it makes no sense to speak about interaction of particles for smaller times; hence, the interval $(t - \delta t; t)$ does not influence the statistical characteristics of the corresponding random processes. The absence of integration in the interval $(t - \delta t; t)$ excludes from consideration the region of singular behavior of integrals for $(t - \tau) \to 0$.

Let us now calculate the one-dimensional probability density function $p(\delta n)$ for fluctuations $\delta n(t)$ using Eq. (36) for the one-dimensional characteristic function. According to the definition [12],

$$p(\delta n) = \frac{1}{2\pi} \int_{-\infty}^{\infty} g_1(\lambda; t) e^{-i\delta n \lambda} d\lambda, \tag{41}$$

that is,

$$p(\delta n) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-\frac{1}{2}\Delta(t)\lambda^2} e^{-i\delta n \lambda} d\lambda. \tag{42}$$

Integrating the last expression, we obtain (see p. 344 of [13])

$$p(\delta n) = \frac{1}{\sqrt{2\pi\Delta(t)}} \exp\left(-\frac{\delta n^2}{2\Delta(t)} \right). \tag{43}$$

Thus, the fluctuations $\delta n(t)$ obey the normal time-dependent Gauss distribution. Plots of the function $p(\delta n)$ versus t are shown in Fig. 4. It can be seen that the probability density $p(\delta n)$ is smeared with time along the δn axis, and at small times, the fluctuations of $\delta n(t)$ are mainly observed at small δn values approaching to the δ-function when $t \to 0$.

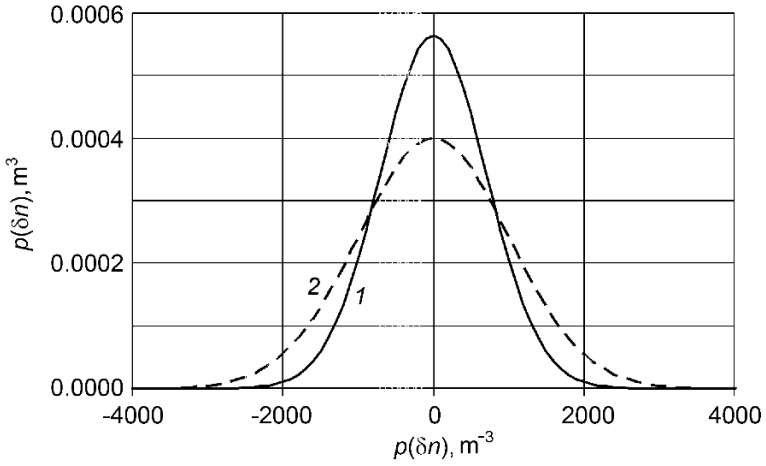

Fig. 4. Plots of the function $p(\delta n)$ for $t = 5\cdot10^{-6}$ (curve *1*) and 1 s (curve *2*).

Taking advantage of Eq. (27), we obtain the spectral density of fluctuations $\delta n(t)$. We derive

$$G_{\delta n}(\omega) = \frac{\sigma}{m^2(D\omega + \beta\sqrt{2D\omega} + \beta^2)}. \tag{44}$$

From the last formula it follows that for $\omega \to 0$, the spectral density $G_{\delta n}(\omega)$ also approaches a constant:

$$G_{\delta n}(\omega)\big|_{\omega\to 0} = \frac{\sigma}{m^2\beta^2} = \frac{\sigma}{m^2\left(\alpha + \dfrac{D}{R_0}\right)^2}. \tag{45}$$

From Eqs. (6) and (27), we derive the expression for the spectral density of particle mass flow $G_{q_m}(\omega)$:

$$G_{q_m}(\omega) = \frac{R_0^2 D\omega + R_0 D\sqrt{2D\omega} + D^2}{R_0^2(D\omega + \beta\sqrt{2D\omega} + \beta^2)}\sigma. \tag{46}$$

Formula (46) demonstrates that the value of the spectral density $G_{q_m}(\omega)$ for $\omega \to 0$ approaches a constant:

$$G_{q_m}(\omega)\big|_{\omega\to 0} = \frac{D^2\sigma}{R_0^2\beta^2} = \frac{D^2\sigma}{(R_0\alpha + D)^2}, \tag{47}$$

and for $\omega \to \infty$, the spectral mass flow density $G_{q_m}(\omega)$ takes the value of the intensity fluctuations of the quantity $\xi_{q_m}(t)$:

$$G_{q_m}(\omega)\big|_{\omega\to\infty} = \sigma. \tag{48}$$

Plots of the corresponding dependences for Eq. (46) for water ($\sigma = 0.5\cdot10^{-8}$ kg^2·s^{-1}·m^{-4} and $\alpha = 11.7$ m/s) and ethyl alcohol ($\sigma = 5.5\cdot10^{-8}$ kg^2·s^{-1}·m^{-4} and $\alpha = 7.4$ m/s) are shown in Fig. 5. Values of α and σ are given by formulas (9) and (10).

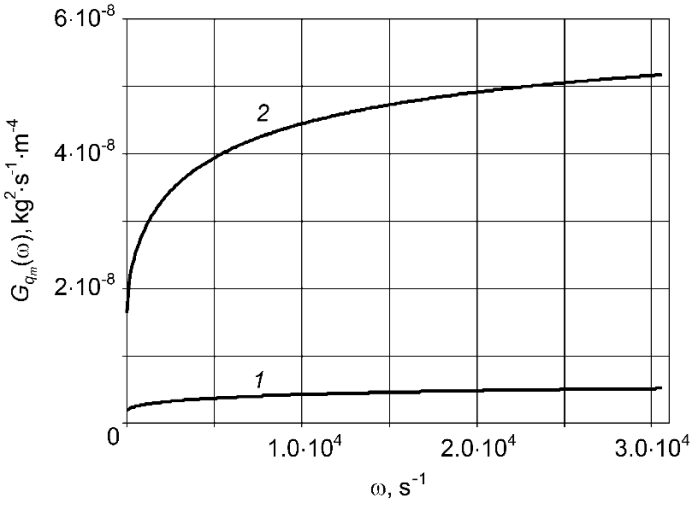

Fig. 5. Plots of the spectral mass flow density $G_{q_m}(\omega)$ corresponding to Eq. (46) for water (curve *1*) and ethyl alcohol (curve *2*).

We note that the statistical characteristics for $\delta n(t)$ and $q_m(t)$ in case of evaporation from the flat liquid surface into the hemisphere are retrieved from the above expressions for $R_0 = \infty$.

SPHERICAL DROP WITH VARIABLE RADIUS

The quasi-stationary case considered in which we neglected changes in the drop radius can be used only as a first approximation. The particle flow through the drop surface causes the mass of the drop and its radius to change. It is especially important to consider changes in the particle radius for small (micron-sized) drops, because their radius can change considerably compared to the initial one even during a small time interval. It is clear that in this case, the quasi-stationary approximation is inapplicable. Changes in the drop radius must also be considered for long-term observations over the behavior of the particle, even for large (several tens and hundreds of microns) aerosol drops. Finally, in calculations of the drop lifetime (up to its vanishing as a result of collapse), consideration of changes in the particle radius is necessary in principle.

However, a solution of the problem on liquid drop evaporation with allowance for changes of the drop radius is a mathematically complex problem (as is well known, the formulated problem belongs to the class of the so-called Stefan problems and analytical solutions are known for a very limited number of cases [14]). In this regard, the statistical characteristics of the physical quantities describing the formulated problem can be calculated only using numerical methods.

In numerical calculations, we took advantage of formulas obtained for the quasi-stationary case with allowance for changes in the drop radius. The change of the drop radius during time *dt* can be found if we know the mass flow $q_m(t)$. Indeed, we obviously have

$$\frac{dR(t)}{dt} = -\frac{q_m(t)}{\rho}, \tag{49}$$

where ρ is the liquid density.

The calculated dependence of the drop radius on the current mass flow through its surface was used in each iteration step when performing numerical calculations. In numerical calculations, we used the following values of the

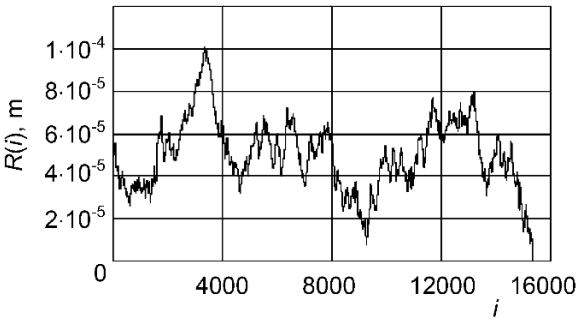

Fig. 6. Typical time changes of the drop radius *R*.

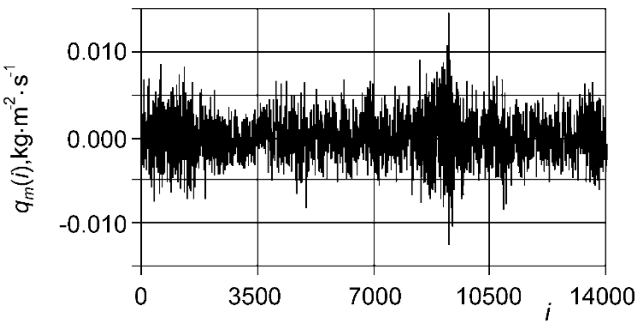

Fig. 7. Typical mass flow q_m depending on time.

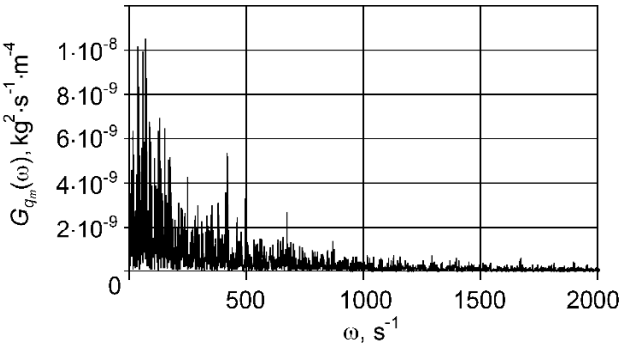

Fig. 8. Spectral mass flow fluctuation density $G_{q_m}(\omega)$.

parameters: the number of iteration steps 30,000, the time step size $\Delta t = 0.1$ s, the mass of the molecules forming the drop $m = 10^{-22}$ kg, the diffusion coefficient $D = 2 \cdot 10^{-5}$ m^2/s, the initial particle radius $R_0 = 50$ μm, the liquid density $\rho = 1000$ kg/m^3, the concentration of saturated vapors $n_0 = 10^{23}$ m^{-3}, and the intensity of the fluctuating component of the mass flow and the accommodation coefficient were calculated from formulas (9) and (10).

Numerical calculations were complicated by the circumstance that the application of the true mass of individual liquid molecule required a very large number of iteration steps up to the drop collapse; this, in turn, required much computation time. In this regard, the mass of the molecule in our calculations exceeded its true value by three orders of magnitude, which formally reduced the number of molecules forming the drop and hence caused its faster evaporation. This allowed us to reduce considerably the computation time and hence to obtain such number of independent realizations which provided sufficient statistics of the results obtained.

It should be noted that overestimation of the mass of the individual molecule causes no principal changes in the character of statistical characteristics of the examined diffusion process and allows the main special features of the model of liquid drop evaporation to be traced with allowance for fluctuations of flow of particles through its surface.

In numerical calculations, we also considered relatively large iteration step in time. This was caused by the necessity of provision of calculation stability with the increase in the iteration step size. However, in this case the statistical characteristics of the examined model did not change essentially. In particular, they retained their form when the time step size was multiply changed.

Figure 6 shows the typical dependence of the particle radius on the iteration step *i*, and Fig. 7 shows the mass flow corresponding to this case.

Figure 8 shows the spectral density $G_{q_m}(\omega)$ of the mass flow shown in Fig. 7. The equation approximating the dependence obtained by the power-law function has the form

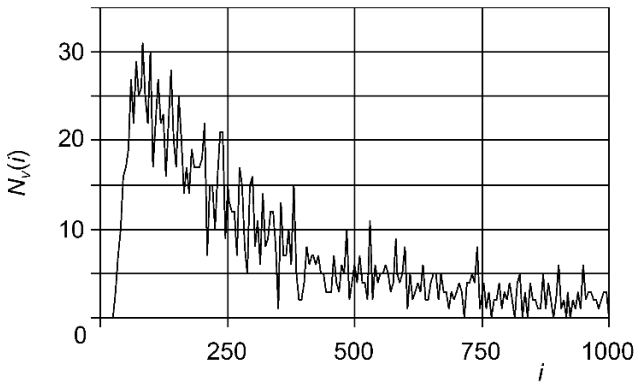

Fig. 9. Distribution of the number of particles $N_v(i)$ disappeared in the *i*th iteration step.

$$G_{q_m}(\omega) = 10^{-7}\omega^{-1.03}. \tag{50}$$

It demonstrates that fluctuations of the mass flow represent flicker noise whose spectral density is inversely proportional to the frequency.

Based on the results of 3000 independent realizations, we calculated the moments of the iterative process for which the drop collapse was observed. Figure 9 shows the distribution of the number of particles $N_v(i)$ disappeared in the *i*th iteration step. It can be seen that the plot is relatively close to that typical of the Poisson process.

The results obtained in this work can be important for investigations of the liquid-drop aerosols presented in the atmosphere. The model suggested in this work considers fluctuations of the mass flow through the boundary between the liquid and gaseous media. It can be used for meteorological investigations, for example, for prediction of the time of fog disappearance, ecological monitoring of pollution of cities and industrial territories by aerosols, investigations of chemical interactions of substances one of which is the aerosol, etc.

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